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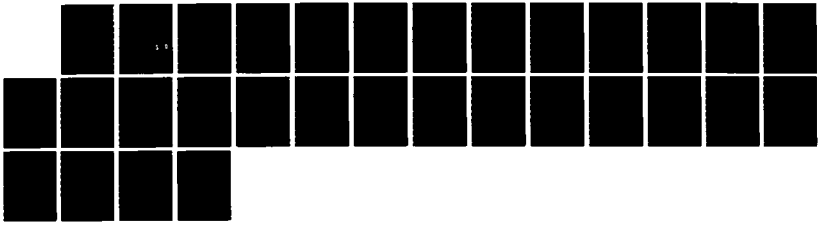

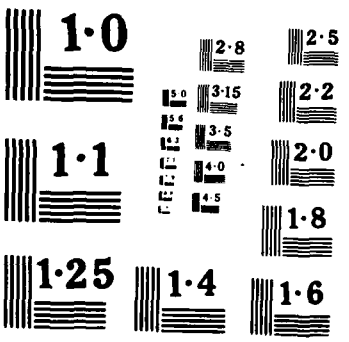
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is appropriate for the time range used in our calculations. The results for different fractal lattices also show that the K-B equation indeed gives a straight line for structure when D/d is not much smaller than unity. As this ratio decreases, deviation from the expected straight line results and an oscillatory behavior is observed. From the oscillatory characteristics, structural information (i.e., fractal dimensionality, and geometrical parameters characterizing the fractal lattices) as well as the molecular interaction parameter γ, characterizing the distance dependence of the exchange interaction, can be determined.

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by

C. L. Yang and M. A. El-Sayed

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University of California
Department of Chemistry and Biochemistry
Los Angeles, California 90024

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Donor-Acceptor One Step Energy Transfer via Exchange Coupling on a Fractal Lattice

C. L. Yang, M. A. El-Sayed*

Department of Chemistry and Biochemistry, University of California at Los Angeles, CA 90024, U. S. A.

ABSTRACT

Temporal behavior of donor intensity, $I_D(t)$, resulting from one step donor-acceptor electronic excitation energy transfer process via exchange mechanism is calculated on fractal lattices, with discrete dilation symmetry, of Euclidean dimension, d = 2, and fractal dimension, D, ranging from 1.99 to 1.0. $I_D(t)$ is fitted to the approximate equation of Klafter-Blumen (K-B) which is useful in fitting experimental results to determine fractal dimension from the slope of the expected straight line obtained by plotting ln (- ln $I_D(t)$) vs. ln ($\ln^D$ t). The result for fractal lattice with D = 1.99 indicates that the approximation is appropriate for the time range used in our calculations. The results for different fractal lattices also show that the K-B equation indeed gives a straight line for structure when D/d is not much smaller than unity. As this ratio decreases, deviation from the expected straight line results and an oscillatory behavior is observed. From the oscillatory characteristics, structural information (i.e., fractal dimensionality, and geometrical parameters characterizing the fractal lattices) as well as the molecular interaction parameter γ, characterizing the distance dependence of the exchange interaction,

can be determined.

INTRODUCTION:

Fractals[1], structures with dilation symmetry, have attracted a great deal of recent attention due to their usefulness in describing disordered systems. Processes occuring in systems such as polymers[2,3], proteins[3,4] and on surfaces[5] have been discussed in terms of fractals, as are processes such as crystal growth[6], dielectric breakdown[7], turbulence and chaos[8]. Fractals have also been used to describe the diffusion of liquids into porous media[9].

Three different dimensions are at least required to define a fractal[10,11]. The first is the Euclidean dimension, d, in which the structure is embedded. The second is called the fractal dimension[1], D. This describes the dependence of the number of sites N(R) on the distance R, through the relation ($N(R) \sim R^D$). The third dimension is the spectral or fracton dimension[10,11], $\tilde{d}$, which governs the random walk and relaxation processes and determines the density of states of the structure. The spectral dimension has been previously discussed in electron-spin relaxation studies in protein[3,4] and triplet-triplet annihilation studies in mixed molecular crystal[12,13].

Recently, studies of the one step electronic energy transfer have been discussed, both theoretically[14] and experimentally[15], in terms of the fractal dimension. Theoretically, Klafter and Blumen[14] (K-B) extended the equation describing the time dependence of a donor intensity derived previously[16,19] to fractal systems for one step energy transfer via dipole-dipole and exchange interaction. The derivations of these equations imply continuous dilation (i.e., the open fractal structure contains holes of any size). In our earlier work[17], we have examined the fit of the calculated time dependent donor intensity for one step dipole-dipole trapping process on the generated fractal

lattice with discrete dilation symmetry. A structure possesses dilation symmetry if it remains equivalent to itself when one changes the unit length by a factor b. If b can be any number larger than one for this structure, the dilation symmetry is continuous. Contrary, if b can only be some discrete values, the dilation symmetry will be called discrete. Structures with discrete dilation symmetry do not possess translation symmetry and contain holes of different sizes as classical fractals. Due to the discrete property, the law, $N(R) \sim R^D$, which governs the varations of the mass N of the structure as function of the distance R, applies on the discrete system only if N(R) is averages over an interval from R to bR. For these reasons, these structures exhibit characteristic microenvironments which are repeated at different length scales. Therefore, calculated time dependent donor intensity plotted in the $\ln -(\ln I(t))$ versus $(\ln t^{D/6})$ showed deviation from the straight line predicted from the K-B equation[14] which is based on the continuous dilation symmetry. The deviation showed an oscillating behavior with a constant periodicity. The amplitude of the oscillation increases as the ratio D/d decreases. Also, from the dilation symmetry criterion, the effect of changing the acceptor concentration is equivalent to a change of time scale for the measurement. This behavior is indeed observed in the plots with different acceptor concentrations, P_A, used in the calculation[17].

In this work, we will examine another kind of interaction, exchange interaction, which is the mechanism reponsible for triplet energy transfer in many aromatic compound systems[20]. The effect of the variation in the microenvironment of the donors on the fractal structures with discrete dilation symmetry is checked for the exchange interaction. The temporal behavior of donor intensity undergoing a one step energy transfer via exchange interaction

on a fractal structure is calculated. Due to the difference in the system for which the K-B equation[14] is derived (systems of continuous dilation), deviation from straight line behavior is expected. We need to find answers to the following questions: a) can we observe the oscillatory behavior for exchange-type transfer similar to that observed for dipolar transfer with the same parameters (i.e., fractal dimension D, acceptor concentration P_A)? b) do the scaling properties of the donor emission intensity that relate to changes in acceptor concentration and of the time scale exist or not in exchange type transfer? c) for exchange interaction, is there a scaling property relating the interaction parameter, γ, (which measure the range of the exchange interaction) and the time scale of the transfer?

The calculation is carried out on several simple fractal structures all having Euclidean dimensionality d = 2 but with fractal dimensionality D = 1.99, 1.75, 1.5, 1.25, and 1.0. The acceptor concentrations P_A and the interaction parameters γ are varied to examine the dilation symmetry criterion. Like the results of dipole-dipole interaction, calculations show that the K-B equation is useful for most structures. A straight line relationship was obtained from which the correct D was determined for structures with D/d not largely different from unity. However, as D/d decreases, oscillatory behavior appears with characteristics relating to the different geometrical parameters describing the fractal structure. The dilation symmetry criterion is then examined and indicates that these properties may give us another approach to determine whether a system has the dilation symmetry property just by varying the acceptor concentration and/or changing the interaction parameter γ by selection of different pairs of donors and acceptors. However, due to the short time intervals and concentration usually employes in experimental studies, the

oscillatory behavior might be difficult to observe.

THE MODEL:

We consider only one step donor to acceptor energy transfer via exchange interaction. Donor-donor energy transfer is excluded. Furthermore, energy levels of the donor and acceptor and the temperature are such that no acceptor to donor back transfer is allowed.

Consider the case of an excited donor located at position $\vec{R}_D$ which is surrounded by a statistical distribution $P_A(\vec{R}_D, \vec{R}_A)$ of acceptors at position $\vec{R}_A$. These acceptors can trap donor excitation with transfer rate W(R) which depends on the relative distance $R = |\vec{R}_A - \vec{R}_D|$ between the acceptor and donor. The decay curve of the donor excitation due to energy transfer follows the equation:

$$I_D(t) = \exp -\left(\int d\vec{R}_A P_A(\vec{R}_D, \vec{R}_A)\ (1-\exp\ (-tW(R)))\right) \qquad (1)$$

where $d\vec{R}_A$ is the volume element over which the interaction is carried out. Equation (1) has been used previously assumming all donor positions are equivalent. For cases in which restricted volume exists, the acceptor distribution function $P_A(\vec{R}_D, \vec{R}_A)$ can be varied for different donor sites. For this reason, averaging over the donor position is necessary in our lattice model. The averaging of equation (1) over the position of the donors with distribution $P_D(\vec{R}_D)$ leads to:

$$I_D(t) = \int P_D(\vec{R}_D)\ d\vec{R}_D \exp -\left(\int d\vec{R}_A P_A(\vec{R}_D, \vec{R}_A)(1 - \exp -tW(R))\right) \qquad (2)$$

The transfer rate W(R) from the donor to the acceptors for exchange interaction is

$$W(R) \sim \exp(-\gamma(d-R)) \tag{3}$$

where γ (interaction parameter) is a measure of the range of the exchange interaction, d is the nearest neighbor distance and R is the relative distance between the donor and the acceptor. Substituting Eq. 3 into Eq. 1, and integrating by parts, Eq. 3 can then be expressed as[16]

$$I_D(t) \sim \exp\left(\int \exp(-X)\ (\ln Z - \ln X)^D\ dX\right) \tag{4}$$

where $X = \exp(\gamma R)$, and $Z = (t/\tau)$.

The term $(\ln Z - \ln X)^D$ can be expanded in power series for non-integer D,

$$(\ln Z - \ln X)^D = \ln^D Z \left(1 - D\left(\frac{\ln X}{\ln Z}\right) + \frac{D(D-1)}{2}\left(\frac{\ln X}{\ln Z}\right)^2 + \ldots\right) \tag{5}$$

and can be approximated as $\ln^D Z$ (the first term) when the remaining terms are much smaller. The approximation is valid only if

$$\frac{\ln Z}{D\ (\ln X)} >> 1, \tag{6}$$

which means the approximation is good when the experimental measurement is carried out at time much longer than the near neighbor transfer time. The donor intensity function can be simplified to[18]

$$\ln I(t) \sim \ln^{D} t \qquad (7)$$

which will be used to check our calculation results.

To get discrete dilation symmetry, we use an idea similar to that used by Fournier in his galaxy model[1]. Details of generating the lattice is described in our earlier paper[17]. Figure 1A shows the effect of the first iteration on the generation pattern. The dilation factor b is determined via the relation[17]:

$$b = N_o^{1/D} \qquad (8)$$

where N_o is the number of points in the generating pattern, and equals to 4 in this calculation. Two examples with two different fractal dimension, D = 1.7 and 1.5, are shown in Fig. 1B, and 1C respectively. One can see, from these two examples, that smaller the D value is, the more open the structure becomes (i.e., with larger holes).

Due to the fact that a donor located near a large hole will have different environment than a donor located away from the hole, one has to average over all the different possible sites. However, in order to avoid the problem arising from the finite size effect of the fractal (due to the finite number of iterations used in the calculation), the donors were randomly distributed on the central area of the lattice, while the acceptors were randomly distributed on the whole fractal structures. The number of sites within the central area is sufficiently large to reveal the geometric characteristic of the discrete fractal structure. Also, as mentioned in the result section, the finite size effect has been examined by the dilation symmetry criterion: a) the calculated time dependent intensity I(t) with $\gamma = a$ can be mapped to the result with $\gamma = na$

when changing the time scale by the relation shown in Eq. 9. b) the calculated I(t) with acceptor concentration, P_A, can also be mapped to the result with P_A/N_o when increasing the transfer rate by $\exp(\gamma b)$ according to Eq. 10. The number of iteraction we use to build the whole fractal is 5, which leads to a total number of acceptor sites of 4096. The number of iterations for the central area is 4 and the corresponding total number of donor points is 1024.

RESULTS:

The time dependent donor intensity function, $I_D(t)$, is calculated by using Eq. (2) at different time t, for different acceptor concentrations, P_A, for different fractal dimensions D, and for different interaction parameters, γ, for exchange interaction. A DEC VAX 11/780 computer is used. Numerical integration is carried out on the generated lattice as described in the model section.

The results of time dependent donor intensity function are plotted in accordance with the equation[14] assuming continuous dilation symmetry (see Figs. 2 and 3). Deviations from the predicted equation due to the effect of 'openness' in the fractal structure as the function of its dimensionality (i.e., the degree of compactness), as well as the effect of interation range are examined. We plot either logarithm of I(t) versus logarithm of t raised to power D or the logarithm of the negative logarithm of I(t) versus logarithm of $\ln^D t$. Either plot should give a straight line as equation (7) predicts.

A: THE D (FRACTAL DIMENSION) DEPENDENCE:

One can see from Figs. 2 and 3 that deviation from the behavior predicted

for the case of continuous dilation symmetry occurs. As the fractal dimension decreases, the deviation becomes larger. These deviations are oscillations which increase with time in Fig. 2, and are of constant amplitude as shown in Fig. 3. The derivation of Eq. 7 has used the assumption of long time behavior to simplify the equation. This approximation can be verified by the result of D = 1.99 where the amplitude of oscillation should be very small, and the deviation from the linearity should come from the long time approximation. In Fig. 2, straight line behavior is observed, which suggests that use of the long time approximation in the calculation is valid.

B: THE EFFECT OF γ (INTERACTION PARAMETER) ON THE TIME DEPENDENCE OF THE DONOR INTENSITY:

The result of the donor intensity decay function for different interaction parameter γ for exchange interaction (i.e., D = 1.0, and P_A = 0.05) is shown in Fig. 4. One can notice the differences between different γ's are the scales of the intensity and time. From the property of dilation symmetry, the calculated intensity of $\gamma = a$ can be mapped to $\gamma = na$ by the transformation:

$$I_D(t,\gamma = a) = I_D(t^{1/n},\gamma = na). \qquad (9)$$

This relation can be used to check the finite size effect of lattice sites (minimum length scale for the finite nearest neighbor distance and maximum length scale for the total number of sites in the lattice, 4096) due to time limitation for the computer calculation. If the number of iterations does not purturb the result, one should expect no change in the time behavior if

multiplying the value of γ by n, and decreasing the time scale by the power of 1/n. This is observed in Fig. 4 (curves A and B), where the value of γ is changed from 5 to 10 and it is equivalent to change the $\ln^D$ t scale to 1/2 of $\ln^D$ t (i.e., 40 to 20 for D =1.0).

C: ACCEPTOR CONCENTRATION AND THE TIME DEPENDENCE OF THE DONOR INTENSITY:

In parts A and B of Figs. 2 and 3, the concentration of acceptor, P_A, is 0.05 and 0.0125, respectively. One can see the difference between them is mainly the scale of the intensity. The shape and the curvature of these two plots are exactly the same (i.e., one can superimpose one plot on the other).

Like Eq. 9 in part B of this section, there is similar dilation symmetry criterion for the relation between time and concentration,

$$I_D(t, P_A) = I_D(t \cdot \exp(\gamma b), P_A/N_o). \qquad (10)$$

where b is the dilation ratio (Eq. 8), and N_o is the number of points in the generating pattern. If the number of iterations does not purturb the result, one should expect that by dividing the acceptor concentration by N_o (N_o is equal to 4 in this generating pattern) and increasing the transfer rate by $\exp(\gamma b)$, to get the same time behavior. This is due to the fact that increasing the concentration of the acceptor by a factor 4 (N_o) and keeping the time scale constant is equivalent to holding the concentration constant (i.e., the same as decreasing the length scale by a factor b), and decreasing the time scale by a factor of $\exp(\gamma b)$. This infers that Figs. 2B and 3B are the short time part of Fig. 2A and 3A.

DISCUSSION:

In our earlier paper[17], we have shown that fractal lattices with discrete dilation symmetry create another kind of excluded volume (i.e., holes) which is characterized by different shapes of generating pattern of the lattice. When these holes become too important to be neglected, deviations from the temporal behavior predicted for fractals with continuous dilation symmetry are observed. In earlier calculations[17] for one step trapping process with dipole-dipole interaction, we have observed that deviation from the equation[14] showed oscillatory behavior with constant periodicity in the $\ln t^{D/6}$ scale. The amplitude of the oscillation depends on the ratio of D/d. The smaller the ratio is, the larger the amplitude would be.

In this work, the effect of interaction type is examined. Since the difference between the dipole-dipole and exchange interactions is the range of transfer process at a given time (i.e., the time dependent function for the transfer rate), we would expect to see the similar oscillatory behavior with a constant periodicity in the $\ln \ln^D t$ scale deviate from the predicted equation derived for continuous dilation symmetry like the dipole-dipole trapping case. As shown in Fig. 3, the oscillation is indeed observed in the calculation. The reason for this oscillatory behavior is the same as that mentioned for the dipol-dipole case,[17] except the x-scale in Fig. 3 has been replaced from $\ln t^{D/6}$ to $\ln \ln^D t$ because of different time dependent function form of the transfer rate for each interaction type. The effect of changing the interaction parameter γ in the exchange interaction is examined. In Eq. 3, we can see that for the same transfer rate W, as the value of γ increases, the value of R ($R = |\vec{R}_D - \vec{R}_A|$) decreases. Therefore, the larger γ is, the shorter the

distance the transfer probes. Because the total number of acceptors within the distance R is proportional to R^D (the relation is exact only when averaged from R to bR as shown in the introduction section), the donor intensity would show smaller decay as γ increases. This behavior is shown in Fig. 4.

As mentioned in the result's section, we have used the dilation symmetry criterion to check our calculation for the finite size effect. The dilation symmetry criterion relates the change of the acceptor concentration to the change of the time scale via Eq. 10. For exchange interaction, there is another criterion that correlates the change of the interaction parameter γ to the change of time scale via Eq. 9. In principle, this means that a knowledge of the time dependence of the donor decay at a given acceptor concentration (or interaction parameter) should enable us to predit the time dependence of another acceptor concentration (or interaction parameter) and/or time scale. Both correlations can be observed in Figs. 2 and 4, where the acceptor concentrations are 0.05 and 0.0125 (for Fig. 2) and the range parameters are 1., 2., 5., and 10. (for Fig. 4). One can see the only difference between them is the scale of the intensity and the time. The shape and the curvature of these plots are exactly the same (i.e., one can superimpose one plot on the other). In other words, the change of the acceptor concentration reveals that Fig. 2A is the short time behavior of Fig. 2B when one changes the time scale by a factor $\exp(\gamma b)$, or Fig. 2A is the magnification of the upper left-corner of Fig. 2B. Similar results are observed for the change in interaction parameter, curve A in Fig. 4 ($\gamma = 10$) is the short time behavior of curve B ($\gamma = 5$) when one change the time scale t to $t^{1/2}$ (or 1/2 in the $\ln^D$ t scale, D = 1 in Fig. 4).

All the properties mentioned above are results of the dilation symmetry of the system under study. Also, information about the structure of the lattice

can be obtained from the oscillatory behavior. For example, the distance between two maxima in the oscillation in Fig. 3A and 3B is 1.39 in the coordinate of $\ln \ln^D t$ for D equal to 1.0). This coordinate is equal to the logarithm of the length scale with power D (i.e., $\ln R^D$). Therefore, the corresponding value of R between two maxima is equal to 4, which is the value of b (i.e., in this case, D = 1.). The change in the logarithm of negative logarithm of the intensity corresponding to this period is equal to $\ln N_o$, where N_o is 4 for the generating pattern we choose. Futhermore, D still can be determined from the slope of the best straight line fit through the oscillating function. Finally, the dilation symmetry criterion can give us another way to examine whether the system investigated possesses dilation symmetry, experimentally, by changing the acceptor concentration and/or interaction parameter for exchange interaction by selecting different donor and acceptor pairs.

In principle, the oscillatory behavior of the deviation from the predicted equation should give us a lot of information about the structure of the system. However, in real systems, due to the limitation of the lifetime and the transfer time of the donor excited state, experimental determination of real behavior is difficult. For example, in triplet-triplet energy transfer between aromatic hydrocarbons (which occurs via the exchange mechanism[20]), the radiative lifetime for the triplet state of these molecules is on the order of tens of seconds. If the transfer rate is too fast, experimentally, it would be difficult to follow the temporal behavior of the energy transfer process. Therefore, only a small time range (i.e., 10^{-4} to few seconds) can be studied experimentally. Assuming that the near distance is d and the transfer time τ within this distance d is $\sim 10^{-12}$sec, then, the distance probed at 10^{-4}sec is about 4.7 d for $\gamma = 5$ (as

was inferred for aromatic molecules in crystals[20]), and the distance probed at 10 secs is 7.0 d. Crude estimation shows that the distance range probed in the triplet-triplet transfer process for most aromatic hydrocarbons would be only about 2.3 d, and it would be very difficult to tell the real structure of the system studied from a simple one step energy transfer process.

The existence of oscillatory behavior indicates that for some types of lattices which follow the equation: $N(R) \sim R^D$, for the fractal structures, it is not necessary that the temporal behavior for trapping process do follow the equation derived from the continuous dilation symmetry. However, the equation would describe the temporal behavior well if the systems considered really possesses the continuous dilation symmetry and the oscillatory behavior would disappear.

CONCLUSIONS:

From the calculation of the temporal behavior of the donor intensity on a fractal lattice with discrete dilation symmetry, the K-B equation is found to describe most of the time dependent behavior for structures with D/d ratios close to unity. In these cases, a plot of ln of negative logarithm intensity versus ln of the logarithm of the time shows a straight line, and the resulting slope gives the value of the fractal dimension D. However, as the D/d ratios decrease, deviations from linearity become large, and oscillatory behavior observed. Fortunately, from the oscillatory behavior, information regarding the geometry of the fractal structure under study can be obtained.

In principle, the results obtained for the cases of small D/d, for different acceptor concentrations and/or for different interaction parameters

should enable experimentalists to test whether the anomalous decay is due to a real fractal space or to a regular structure with excluded volume. For fractal structure, it is possible, at least in principle, to characterize the fractal dilation symmetry experimentally by determining the value of N_o and b from the oscillatory, temporal behavior of donor intensity. Unfortunately, the ranges of time and concentration which can be studied experimentally in energy transfer processes do not allow observing full oscillation. Instead, a part of the oscillation could only introduce errors in equation (7) when used to determine D. Furthermore, the value of D determined could depend on the time interval plotted (i.e., on different portion of the oscillation used). Thus, due to the experimental limitation imposed by the lifetime of the donor excited state, and the short range of the transfer of the exchange interaction mechanism, the determination of the real fractal dimension from one step exchange energy transfer might be met with uncertainty.

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Figure captions:

Fig. 1:

(A) Plot of the generating function of the fractal lattice (N_o = 4) used in this study. The value of b for different D's are calculated by Eq. 9. (B) and (c) are the two typical fractal lattice structures obtained with this generating function for D=1.7 (scaling factor b = 2.26) (B), and 1.5 (scaling factor b = 2.52) (C).

Fig. 2:

Plot of the calculated ln of the donor intensity versus $\ln^D t$ for different fractal dimension D, and for different acceptor concentration P_A. — — — D=1.99, —·—·— D=1.75, — . — D=1.5, — — — D=1.25, - - - - D=1.0. In part A, and B, the acceptor concentration is 0.05 and 0.0125, respectively. t is in arbitrary unit. The characteristic transfer time has been kept constant in Fig. A and B, but are different for each concentration.

Fig. 3:

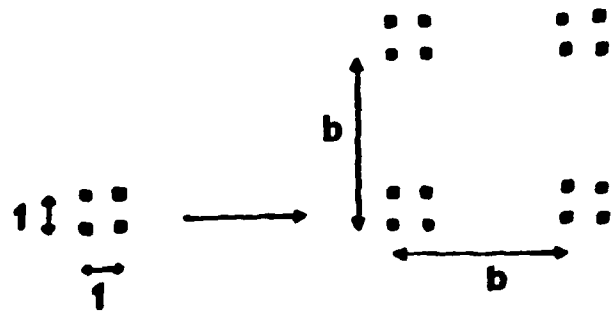
Plot of the ln(-(ln(intensity)) versus ln $\ln^D$ (time) for different fractal dimension D and for different acceptor concentrations. The slope of the curves

should be equal to 1 in these coordinates if the fractals have continuous dilation symmetry assumed by ref. 14. The slopes calculated are oscillating with time.——— D=1.99, —··— D=1.75, —·— D=1.5, — — — D=1.25, - - - - D=1.0. (A) and (B) correspond to acceptor concentrations of P_A=0.05, and 0.0125 respectively. t is in arbitrary unit. The best straight lines fit through all are parallel to each other.

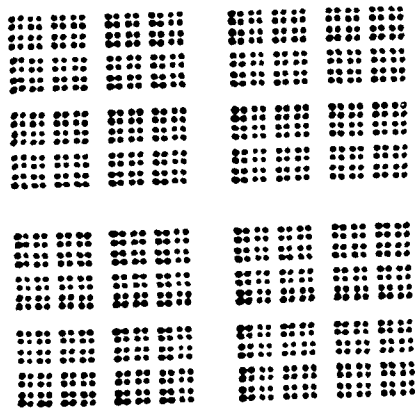
Fig. 4:

Plot of the ln (intensity) versus $\ln^D$ (time) for different interaction parameters γ, A: γ=10., B: γ=5., C: γ=2., D: γ=1. All the data calculated are with the same fractal dimension D (D=1.0) and acceptor concentration (P_A=0.05). t (time) is in arbitrary unit, and is the same for all four curves. The curve A can be mapped to curve B by changing the time scale via Eq. 9 due to the dilation symmetry property of the generated lattice. The same procedure can also be applied to curve B, C, and D.

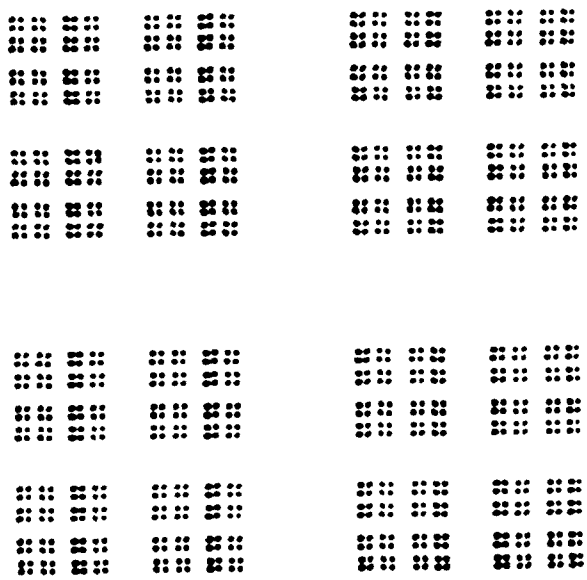
A

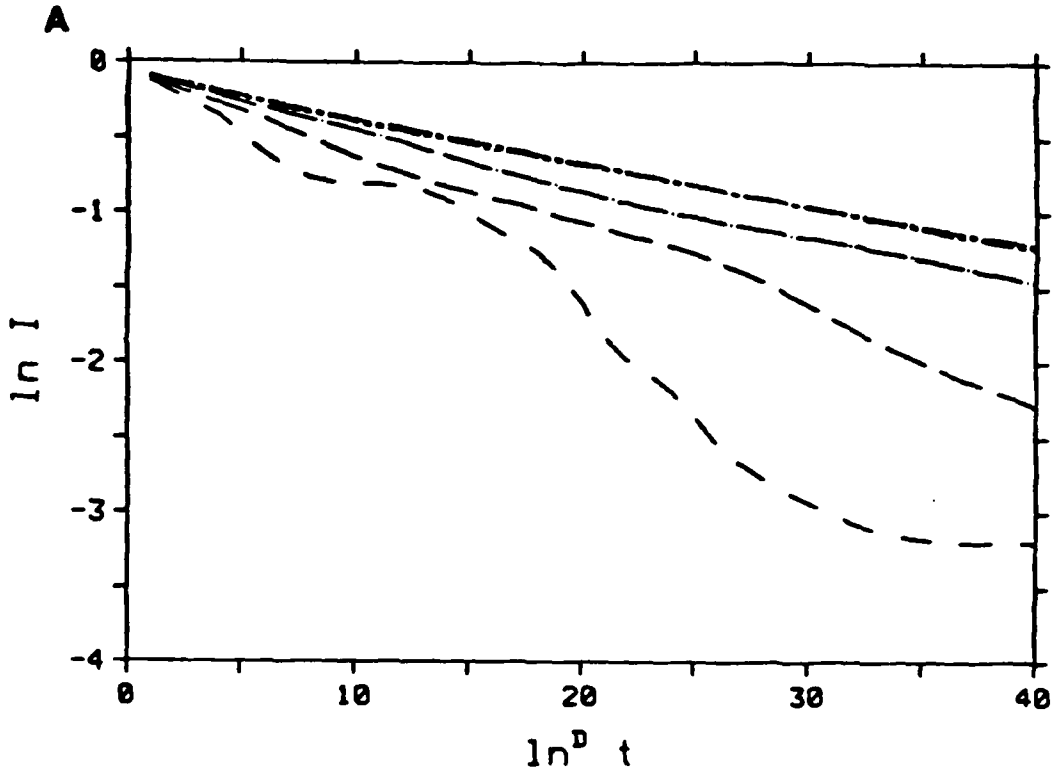


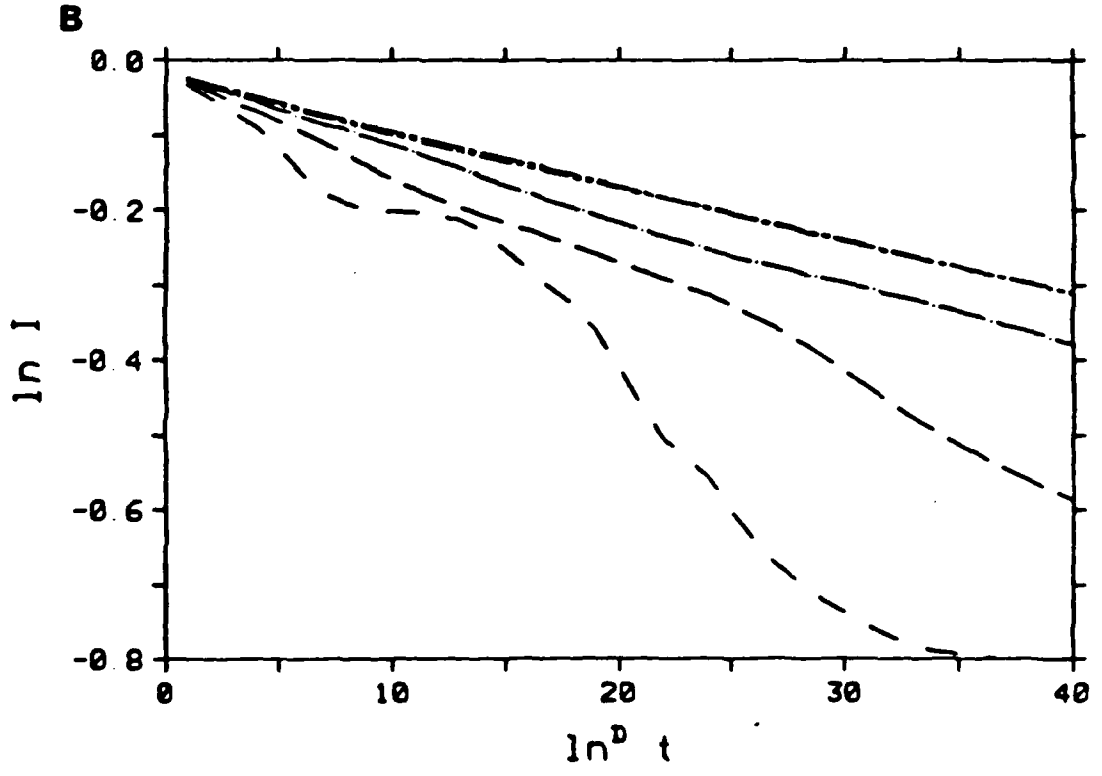
B



C




A
0
-1
-2
-3
-4
ln I
0
10
20
30
40
ln^D t


B
0.0
-0.2
-0.4
-0.6
-0.8
ln I
0
10
20
30
40
ln^D t

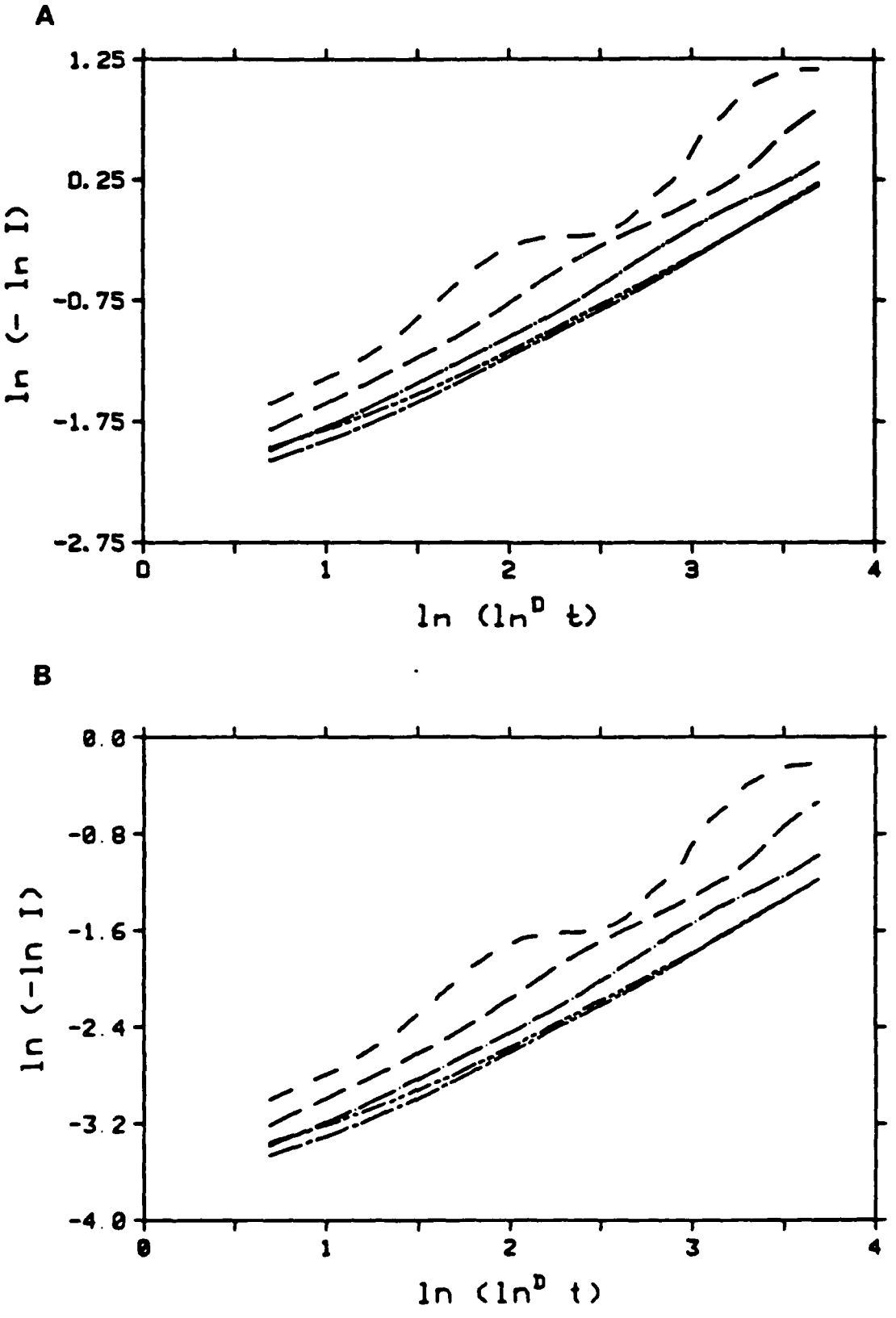

A
1.25
0.25
-0.75
-1.75
-2.75
ln (- ln I)
0
1
2
3
4
ln (ln^D t)
B
0.0
-0.8
-1.6
-2.4
-3.2
-4.0
ln (-ln I)
0
1
2
3
4
ln (ln^D t)

Fig 3

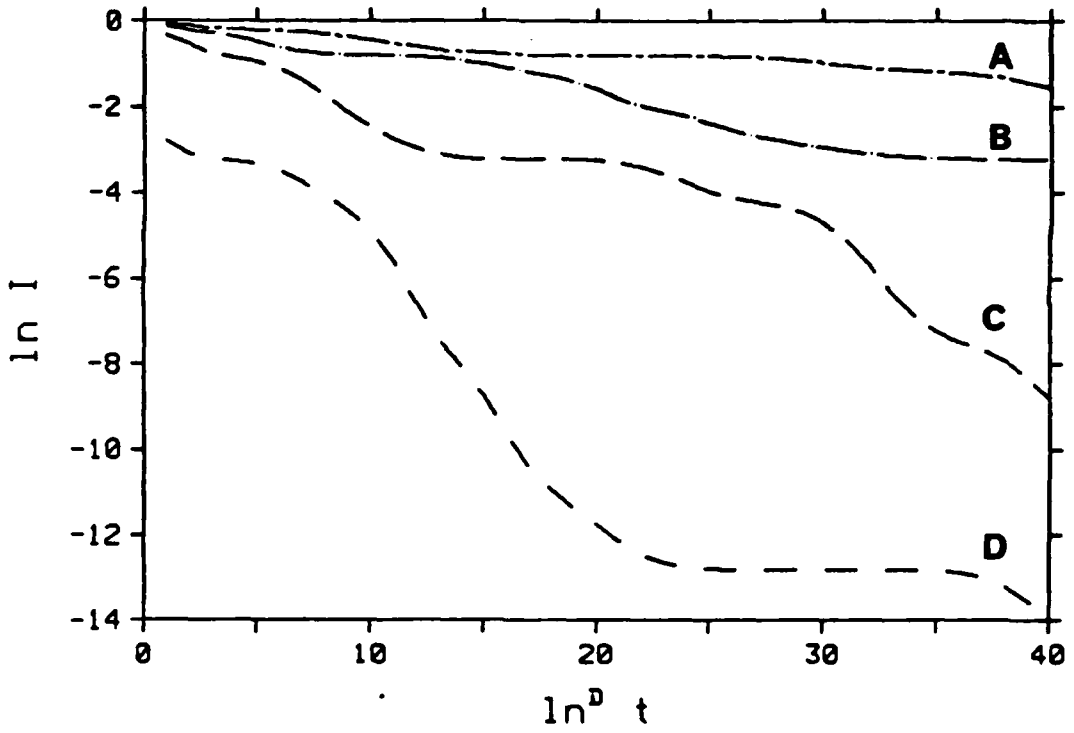


Fig -

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